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HIGH POWER UV LASER PUMPED DYE LASER TESTS

FINAL REPORT

March 1977

Prepared by

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Advanced Laser Research Laboratory

For

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Photochemical stability of ten different dyes emitting in the blue-green spectral region was investigated, both for KrF and XeF laser pumping, to determine the possibility of developing high power, uv laser pumped dye lasers. Several dyes were found to exhibit better stability under uv laser pumping in comparison to flash lamp pumping. Some possible explanations are suggested.

HIGH POWER UV LASER PUMPED DYE LASER TESTS

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1.0 INTRODUCTION AND SUMMARY

Dye lasers in the blue-green spectral region are of great interest for underwater communications and for isotope separation. So far, high power operation of dye lasers has only been reported using flash lamp pumping. Because of geometrical constraints limiting the heat transfer capability, this method of pumping severely restricts the scalability. Furthermore, for some applications high repetition rates are required which cannot be obtained using flash lamps.

These problems can be eliminated by using a high power laser pump along with the jet stream heat transfer technique. The rare gas halide lasers and lamps developed recently are very attractive candidates for a high power dye pump, but the photochemical stability of the dyes under the intense near uv emission of the rare gas halide lasers has been unknown. This program was specifically designed to address this photochemical stability issue.

During the course of the program, ten different dyes having emission in the blue-green region were investigated with both KrF and XeF radiation as pump sources. Commercially available as well as dyes specially tailored for photochemical stability were included in the list of candidates The results of this investigation are presented indicating that some dyes actually exhibit better stability with laser pumping than that reported with flash lamp pumping. The results also include the dependency of the photochemical stability and the laser efficiency on pump intensity.

2.0 EXPERIMENTAL ARRANGEMENT AND PROCEDURES

The experimental arrangement is shown schematically in Figure 1. It consists of a dye laser which was transversely pumped by the KrF/XeF laser, and detection equipment. A Molectron cuvette dye cell Model DL051 with magnetic stirrer was used. This cell is A.R. coated and has a useful volume with a diameter of 0.8 cm and a height of 5 cm. It was filled with approximately 0.8 cc of the dye studied, and the dye was stirred between shots. Various output couplers were tried, but for almost all cases a coupling of 50% was found to be the optimum (couplers with reflectivities of 90%, 80% and 50% were available). Lenses L_1 and L_2 focused the pump beam to an approximately elliptical spot with dimensions 0.8 cm x 0.3 cm. Two Gen-Tec pyroelectric detectors with sensitivities of approximately 1 mV/mJ were used to measure the uv energy and the dye laser energy. Because of the low repetition rate, approximately one pulse per minute, the outputs of the two detectors were displayed on the screen of a storage oscilloscope, measured, and individually recorded.

Longitudinal pumping was also attempted. However, because of the high uv intensities needed to pump the dye, pumping through the dye laser mirrors was impossible and the pump beam had to be introduced at a small angle instead.[1] Due to its large divergence (10 mrad at the output of the laser) the beam could only be collimated over a very short length, however, such that the angle between the dye laser optical axis and the pump beam axis had to be relatively large. This resulted in poor coupling to the dye laser cavity and thus in low efficiencies. For this reason, and because it did not interfere with the real goal of this program, transversal pumping was employed in all the tests. A disadvantage of transversal pumping compared to longitudinal pumping is that, because of the spatially inhomogeneous gain (decreasing away from the input window), energy extraction is not complete. This results in lower efficiencies.

1. D. J. Bradley, G. M. Gale, M. Moore, and P. D. Smith, "Longitudinally Pumped, Narrow-Band Continuously Tunable Dye Laser," Phys. Letters, Vol. 26A, 11 March 1968.

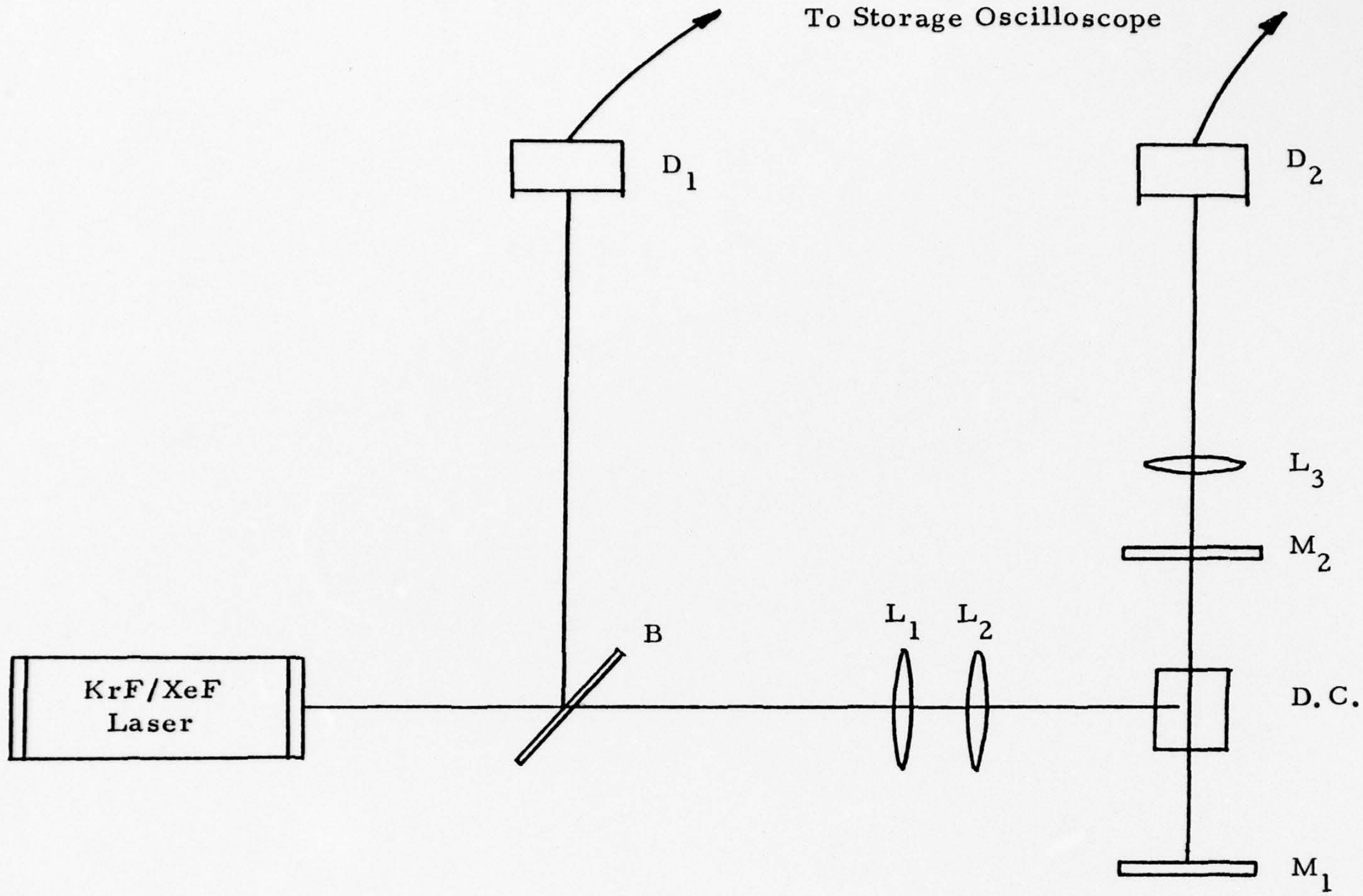

Figure 1. Experimental setup for dye testing. B = beamsplitter; D_1, D_2 = pyroelectric detectors; D. C. = dye cell; M_1, M_2 = dye laser mirrors; L_1, L_2 = lenses to focus the pump beam; L_3 = lens to decrease the visible beam intensity at D_2.

In almost all cases the dye laser efficiency decreased linearly with the total uv energy deposited, or, for an approximately constant pump energy per shot, with the number of shots. Thus

$$\mathcal{E} = \mathcal{E}_{max} - \frac{\partial \mathcal{E}}{\partial N} N \qquad (1)$$

where $\mathcal{E}$ is the efficiency and N the number of shots. Because of the low repetition rate and the high cost of running these experiments due to mirror damage and gas requirements, the total number of shots for a particular dye was chosen such that the efficiency had dropped between 10% and 20% at the end of the test. Only for the case of the dye C102 pumped by KrF was the test continued to the 50% point. Assuming a linear relation between efficiency and number of shots, one finds immediately

$$N_{1/2} = \frac{\mathcal{E}_{max}}{2\,(\Delta\mathcal{E}/\Delta N)} \qquad (2)$$

where $N_{1/2}$ is the number of shots required to decrease the efficiency by a factor of two, and $\Delta\mathcal{E}/\Delta N$ is the slope of the measured $\mathcal{E}(N)$ curve. In a few cases the function $\mathcal{E}(N)$ was not exactly linear. These cases are discussed in more detail in the next section.

2.1 Experimental Results and Discussion

The dyes investigated are listed in Table I, both for KrF and XeF pumping. The experimentally measured curves of efficiency versus number of shots are shown in Figures 2 to 18. In these figures, each data point corresponds to an average over the number of shots obtainable from one gas filling (typically 10 to 20 shots). The values of $N_{1/2}$ calculated using expression (2) are shown in Tables II and III, respectively for KrF and XeF pumping. Also given in these tables is an approximate value for the energy conversion efficiency. It is worth noting that for these uv to visible converters the maximum energy conversion efficiency possible is about 50% for KrF pumping

and 70% for XeF pumping. The efficiencies listed in these tables are by no means to be considered as the maximum values obtainable. In most cases, the efficiency was found to increase with concentration as observed by changing the concentration from 10^{-3} to 6×10^{-4}. As a result, it is not even meaningful to compare the efficiencies for different dyes, since some were more optimized than others. Additional reasons for the fairly low efficiencies are the transversal pumping configuration and the fact that a careful optimization of pumping intensity, etc., was not possible at the low repetition rates used in these experiments.

Due to severe shot-to-shot fluctuation problems for the case of XeF pumping using the coaxial laser (τ = 100 ns), the last three dyes, i.e., C6H, AC2F, and AC3F, were pumped using a uv preionized self-sustained transverse discharge laser (τ = 50 ns). For these three dyes, and also for C102 pumped by XeF, it is very hard to give values for $N_{1/2}$. For C102, the decrease in efficiency after 250 shots at 50 mJ per shot is very small, making it impossible to realistically extrapolate to the 50% point. The value for $N_{1/2}$ shown in Table III was calculated using the slope shown in Figure 14. Because of the good stability of this dye under XeF pumping, it should be studied in more detail. The dyes AC2F and AC3F show a strange $\mathcal{E}(N)$ behavior which is hard to approximate by a single straight line. However, the data clearly indicates that even after 600 shots at 60 mJ/shot, these dyes still showed only a small decrease in efficiency. The values for $N_{1/2}$ shown in Table III for AC2F and AC3F were calculated by adding the number of shots in the plateau region to the number required to decrease the efficiency from its value in the plateau region to 50% of that value (using expression (2)). Clearly, these numbers are preliminary and should be used with caution. Finally, Table IV summarizes the values for $N_{1/2}$ and the conversion efficiency, both for KrF and XeF pumping, scaled to a uv pulse energy of 100 mJ.

Table I. List of Dyes Investigated with KrF and XeF Pumping

Dye	KrF	XeF
C102	x	x
C30	x	-
C1H	x	-
C2H	x	x
C6H	x	x
C6F	x	-
C8F	x	-
Q6F/A	x	-
AC2F	x	x
AC3F	x	x

x = Investigated
\- = Not investigated

Table II. Stability with KrF Pumping

Dye	Concentration	E_{in} (mJ)	$N_{1/2}$	Efficiency (%)
C102 (a)	10^{-3}	35	600	14
(b)	6×10^{-4}	90	250	11
C30	10^{-3}	90	235	6.5
C1H	10^{-3}	90	200	9.5
C2H	10^{-3}	90	185	11
C6H	10^{-3}	90	400	15
C6F	10^{-3}	90	260	6.5
C8F	10^{-3}	90	240	8.5
Q6F/A	10^{-3}	90	215	7
AC2F (a)	10^{-3}	90	770	9
(b)	10^{-3}	90	810	9
AC3F	10^{-3}	90	300	10

Table III. Stability with XeF Pumping

Dye	Concentration	E_{in} (mJ)	$N_{1/2}$	Efficiency (%)
C102	10^{-3}	50	1800*	13
C2H	10^{-3}	50	235	8
C6H	10^{-3}	50	1000*	18
AC2F	10^{-3}	60	1300*	15
AC3F	10^{-3}	60	1700*	16

* See comments in Section 2.1

Note: The quantum efficiency for XeF pumping is about 1.4 times that of KrF pumping.

Table IV. Summary of Stability Results at 100 mJ Pumping

	$N_{1/2}$		Efficiency (%)	
Dye	KrF	XeF	KrF	XeF
C102	220	900	14	13
C30	210	-	6	-
C1H	180	-	9.5	-
C2H	165	120	11	8
C6H	360	500	15	18
C6F	235	-	6.5	-
C8F	215	-	8.5	-
Q6F/A	195	-	7	-
AC2F	710	780	9	15
AC3F	270	1000	10	16

Note: The quantum efficiency for XeF pumping is about 1.4 times that for KrF pumping.

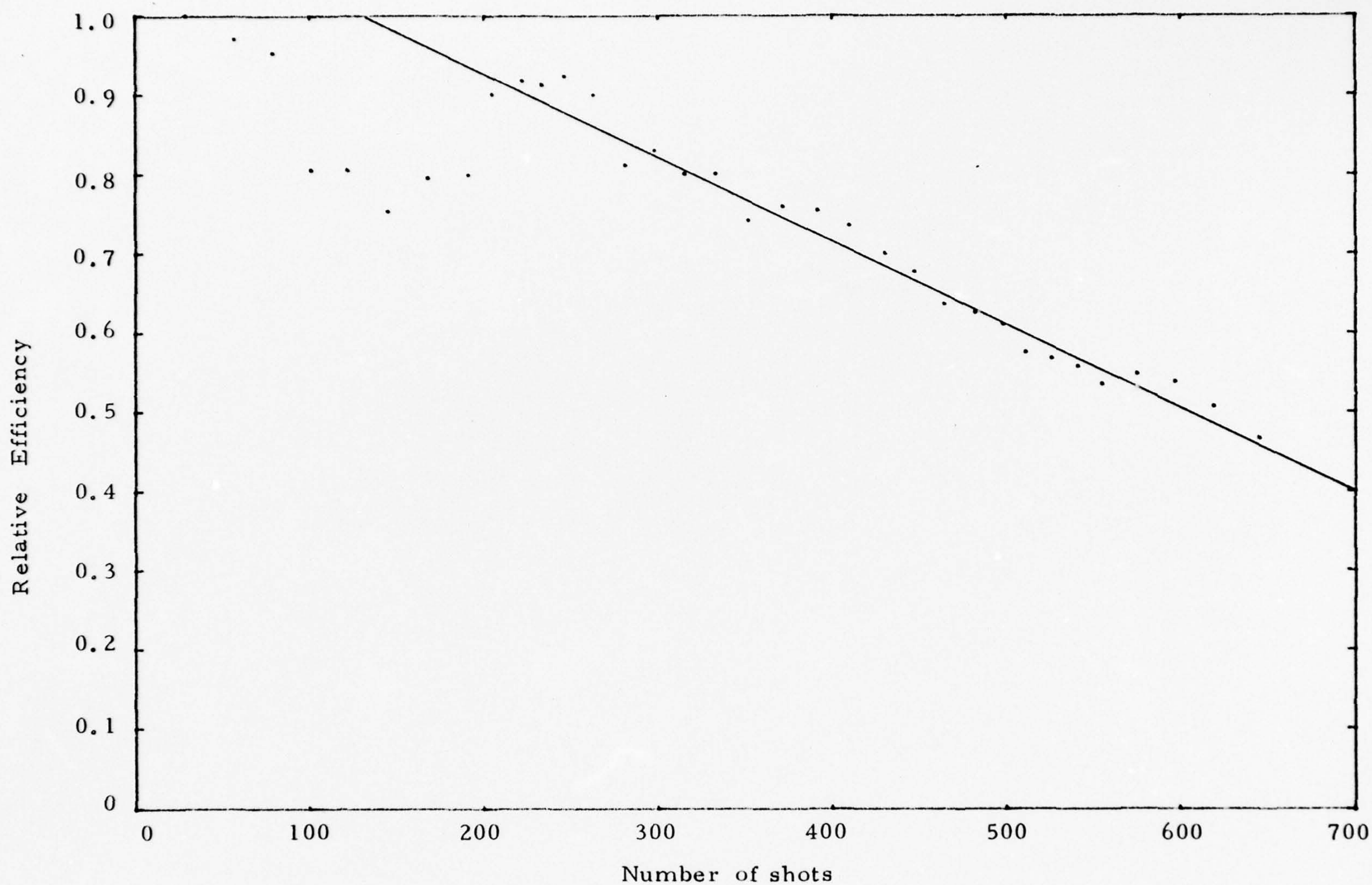

Figure 2. Stability of C102 with KrF pumping. $E_{in} = 35 mJ/shot$

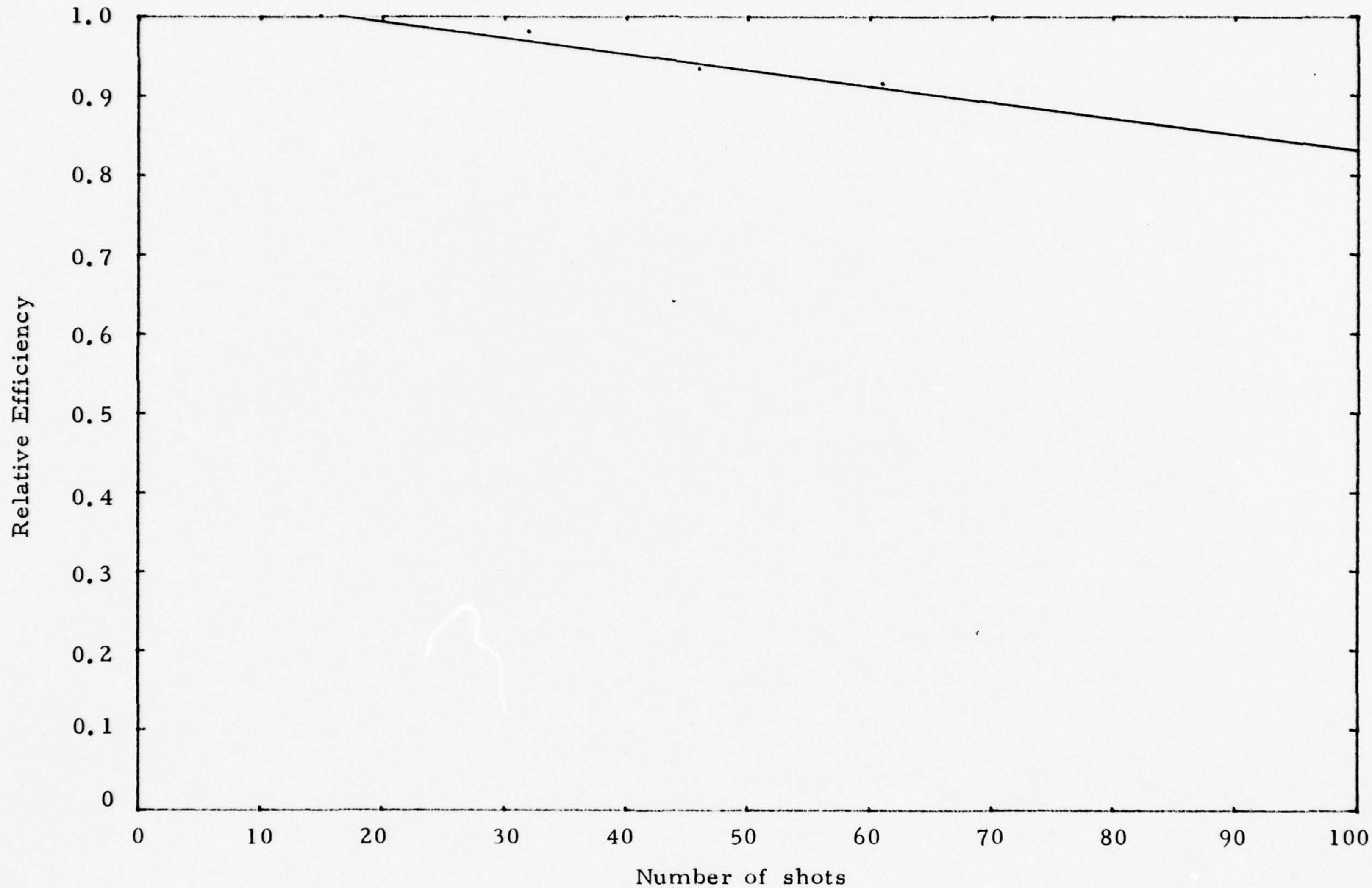

Figure 3. Stability of C102 with KrF pumping. $E_{in} = 90$ m J/shot

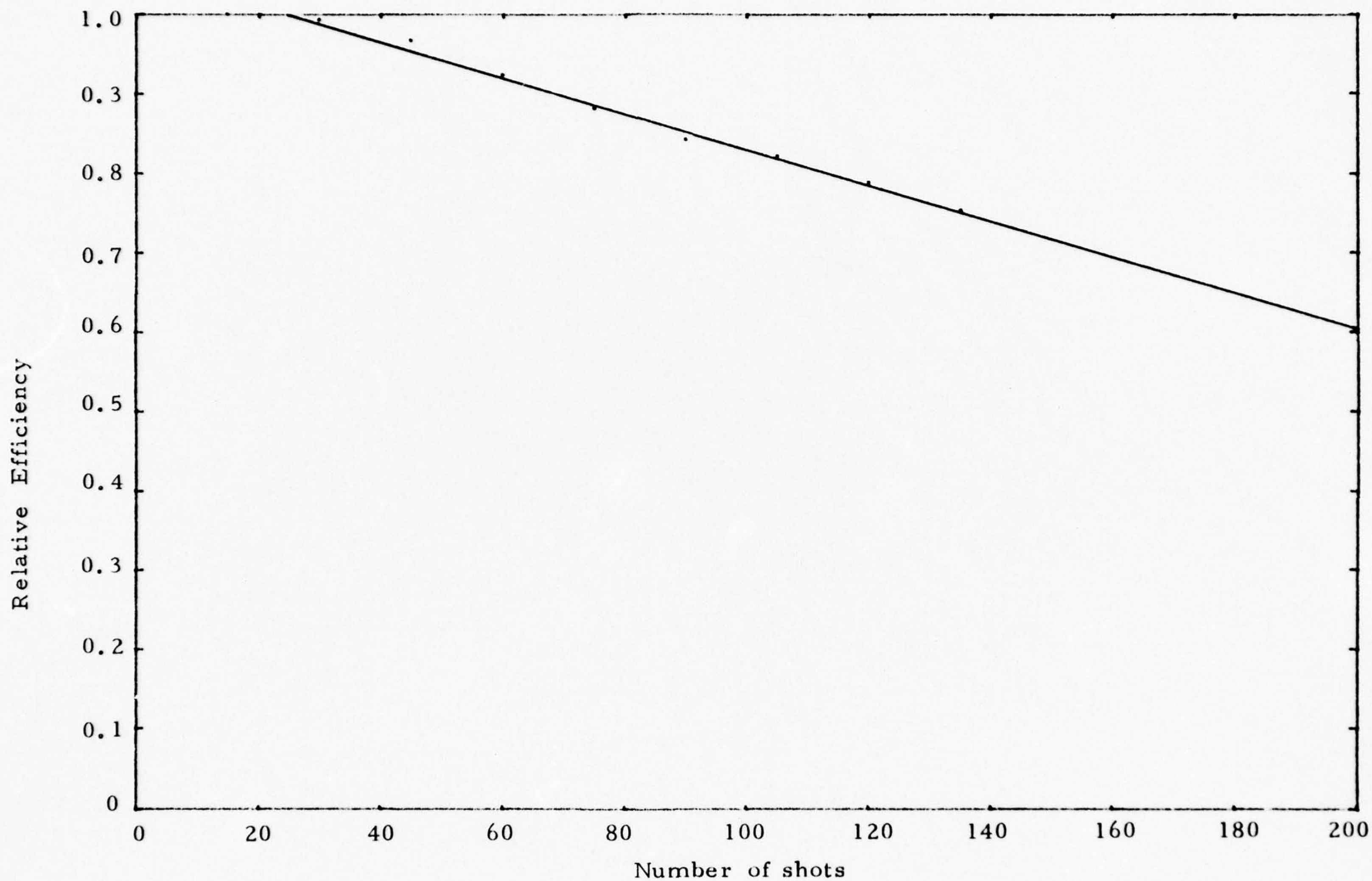

Figure 4. Stability of C30 with KrF pumping. $E_{in} = 90\,mJ/shot$

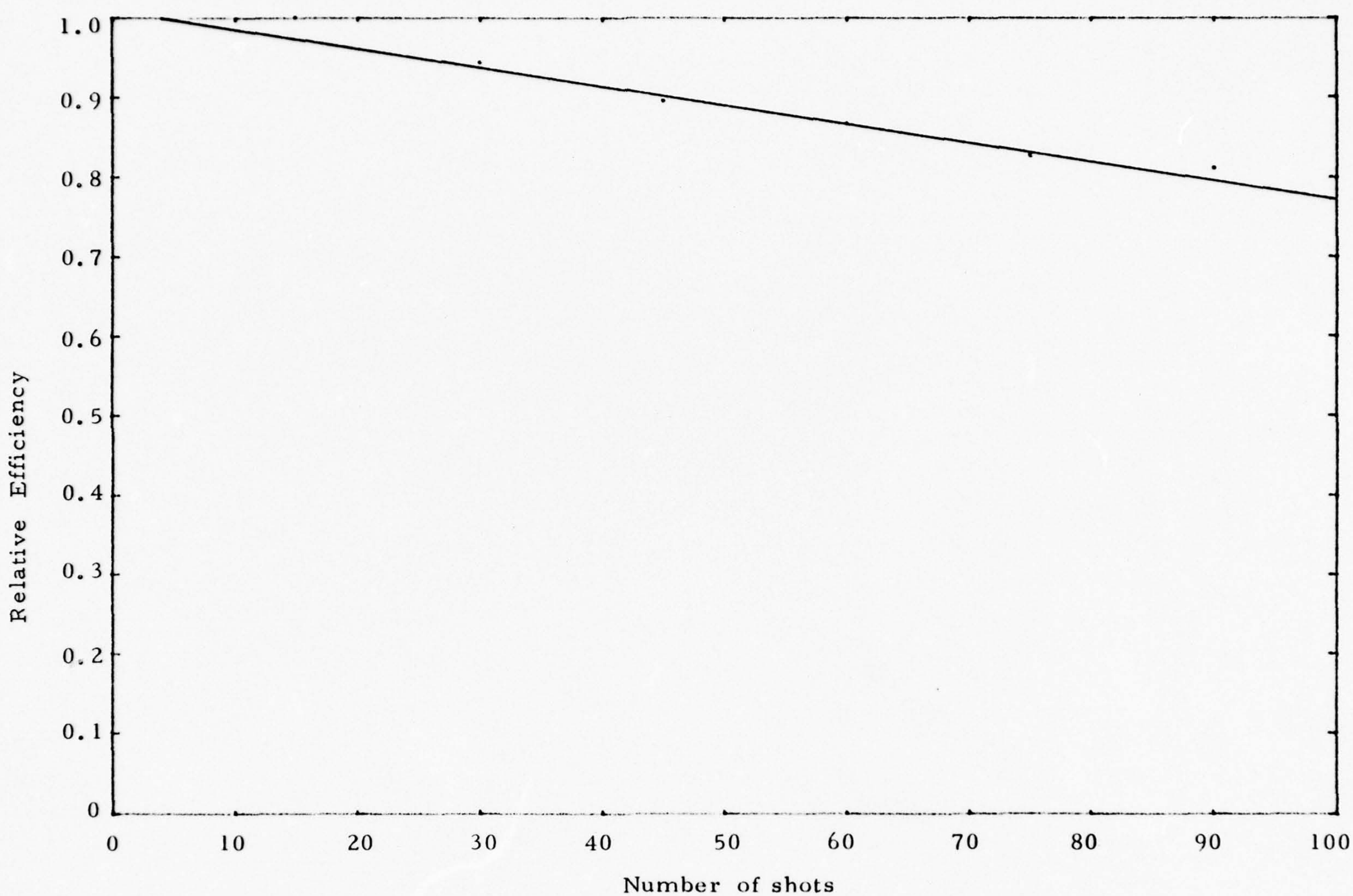

Figure 5. Stability of ClH with KrF pumping. E_{in} = 90mJ/shot.

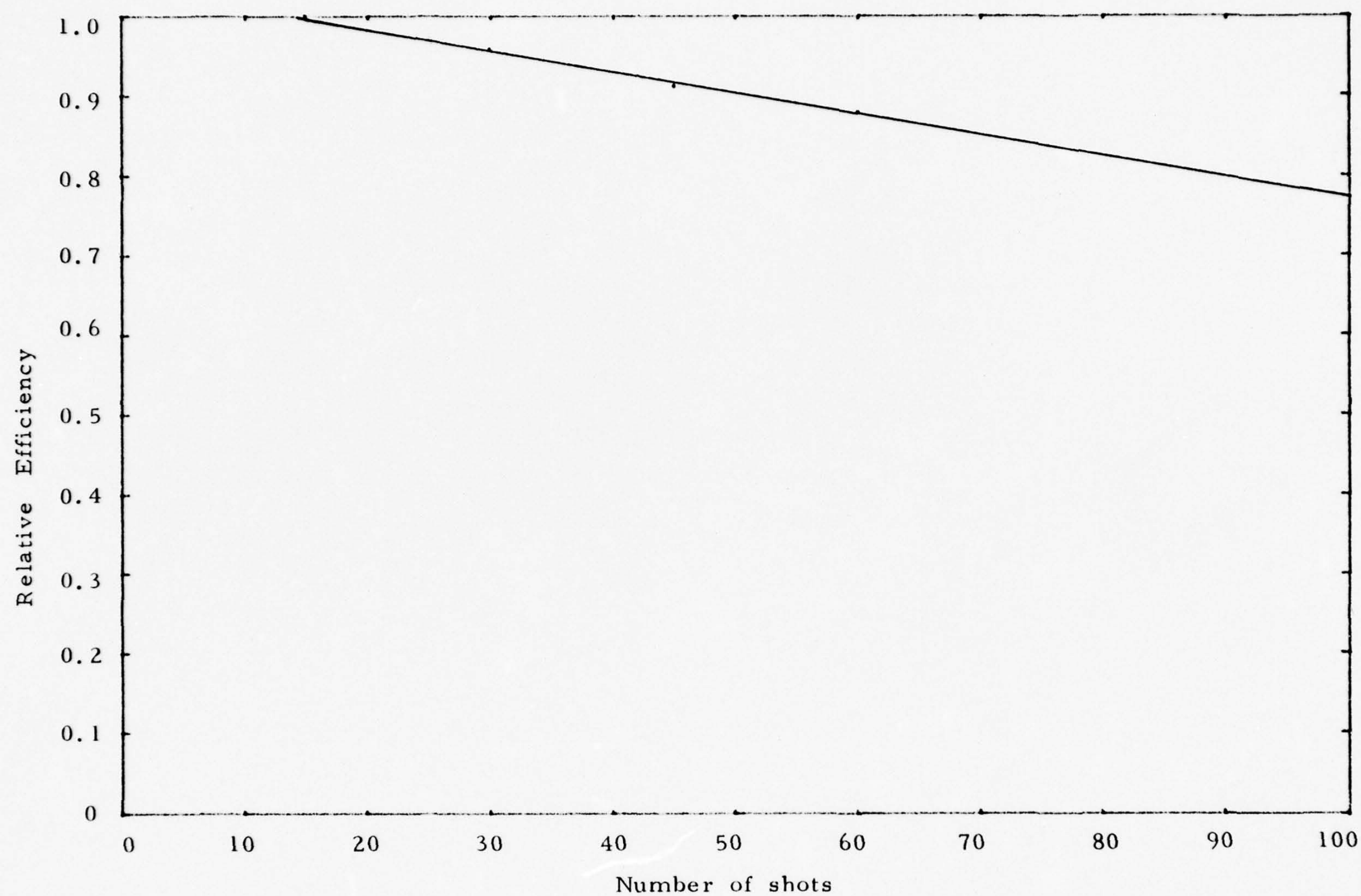

Figure 6. Stability of C2H with KrF pumping. E_{in} = 90 mJ/shot

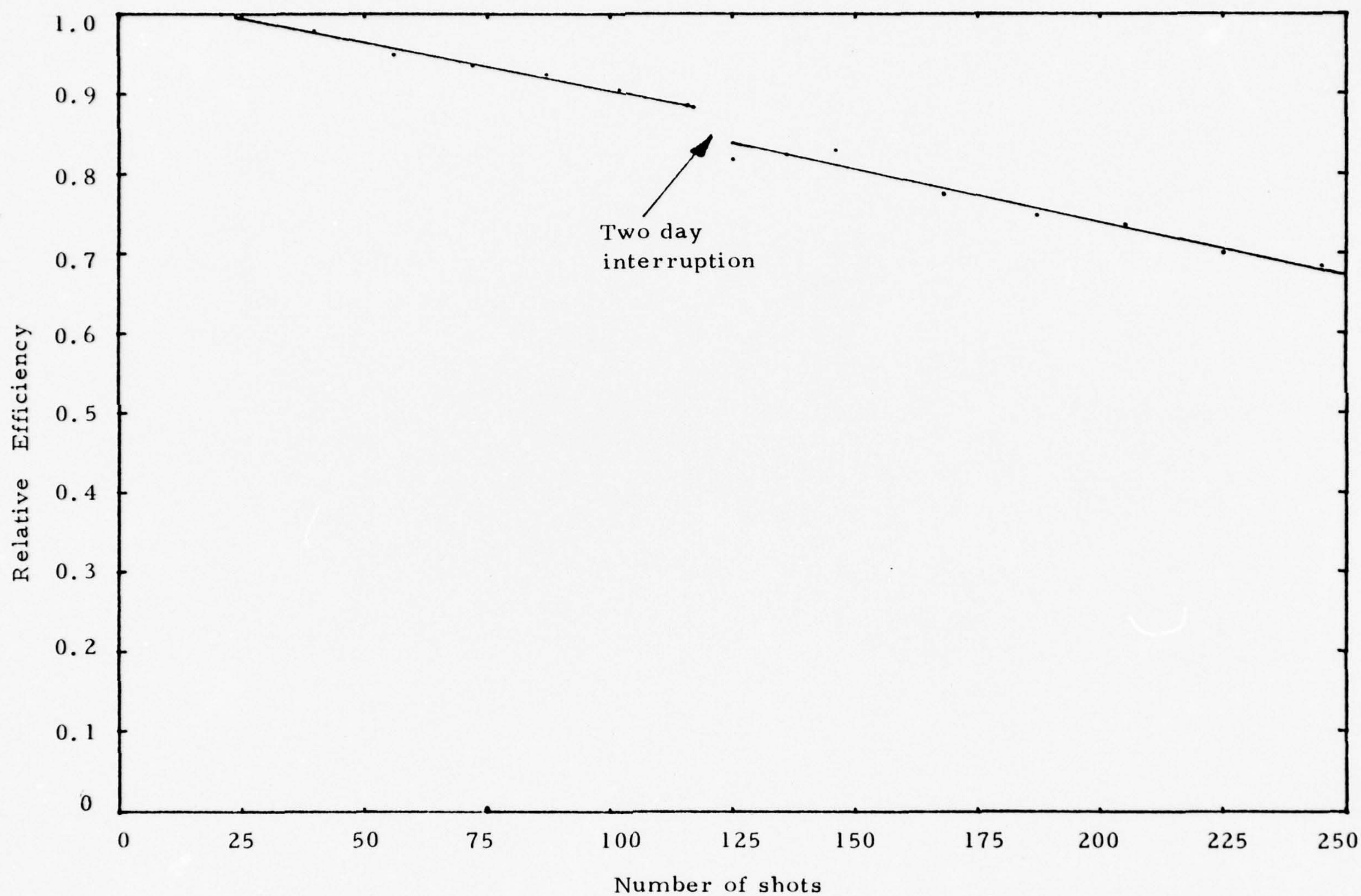

Figure 7. Stability of C6H with KrF pumping. E_{in} = 90 mJ/shot

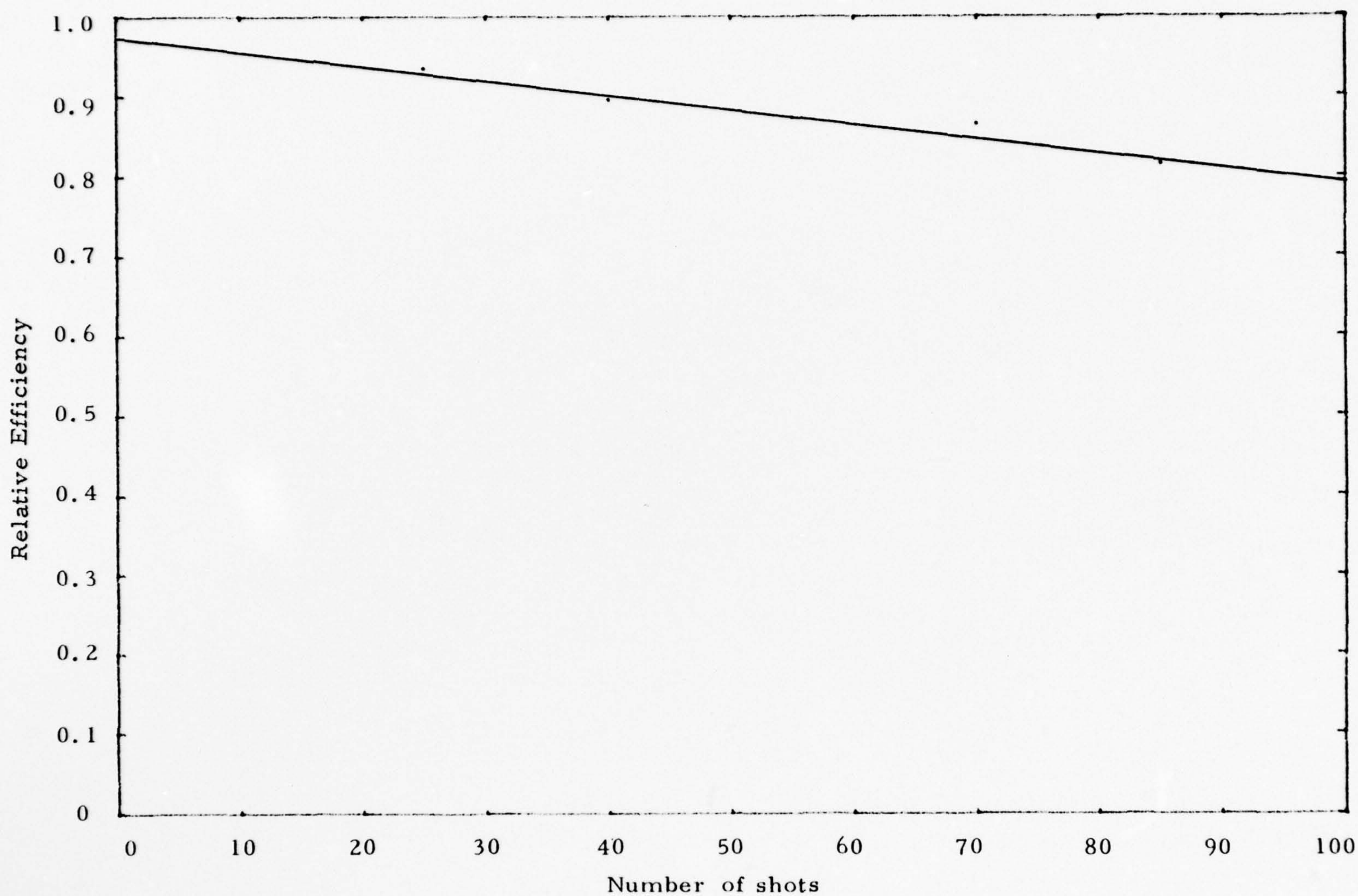

Figure 8. Stability of C6F with KrF pumping. E_{in} = 90 mJ shot

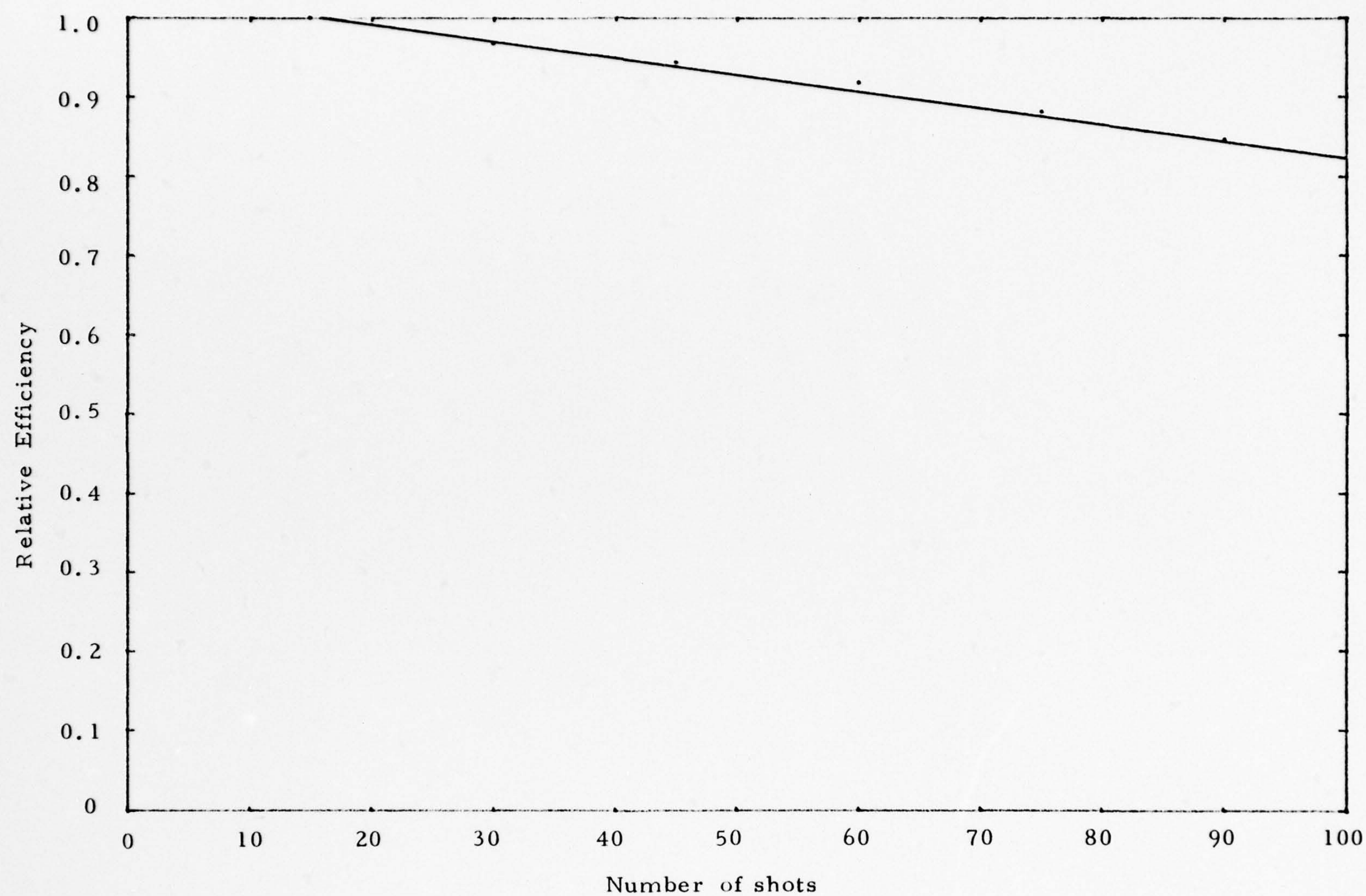

Figure 9. Stability of C8F with KrF pumping. $E_{in} = 90$mJ/shot

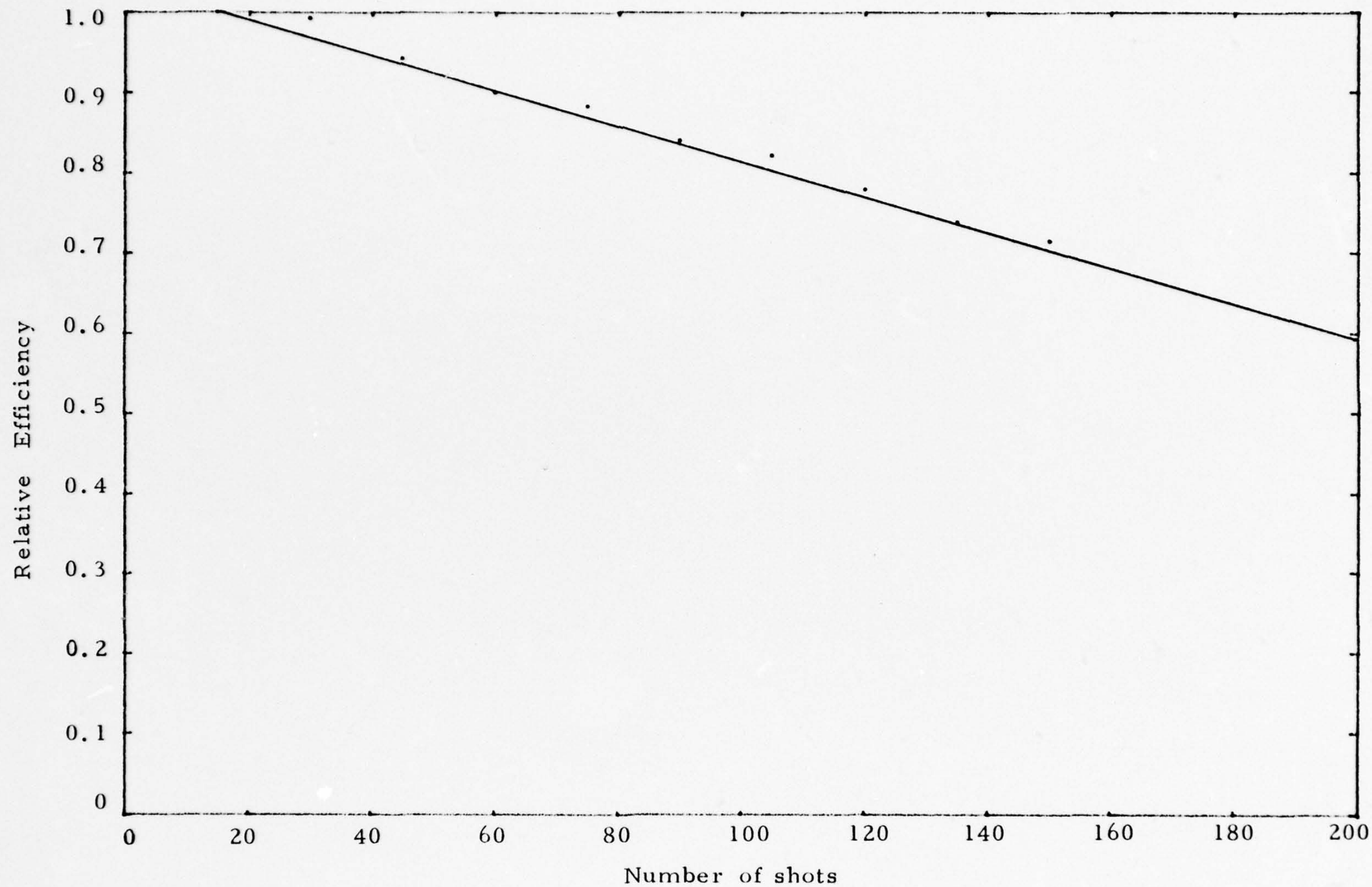

Figure 10. Stability of Q6F/A with KrF pumping. E_{in} = 90mJ/shot

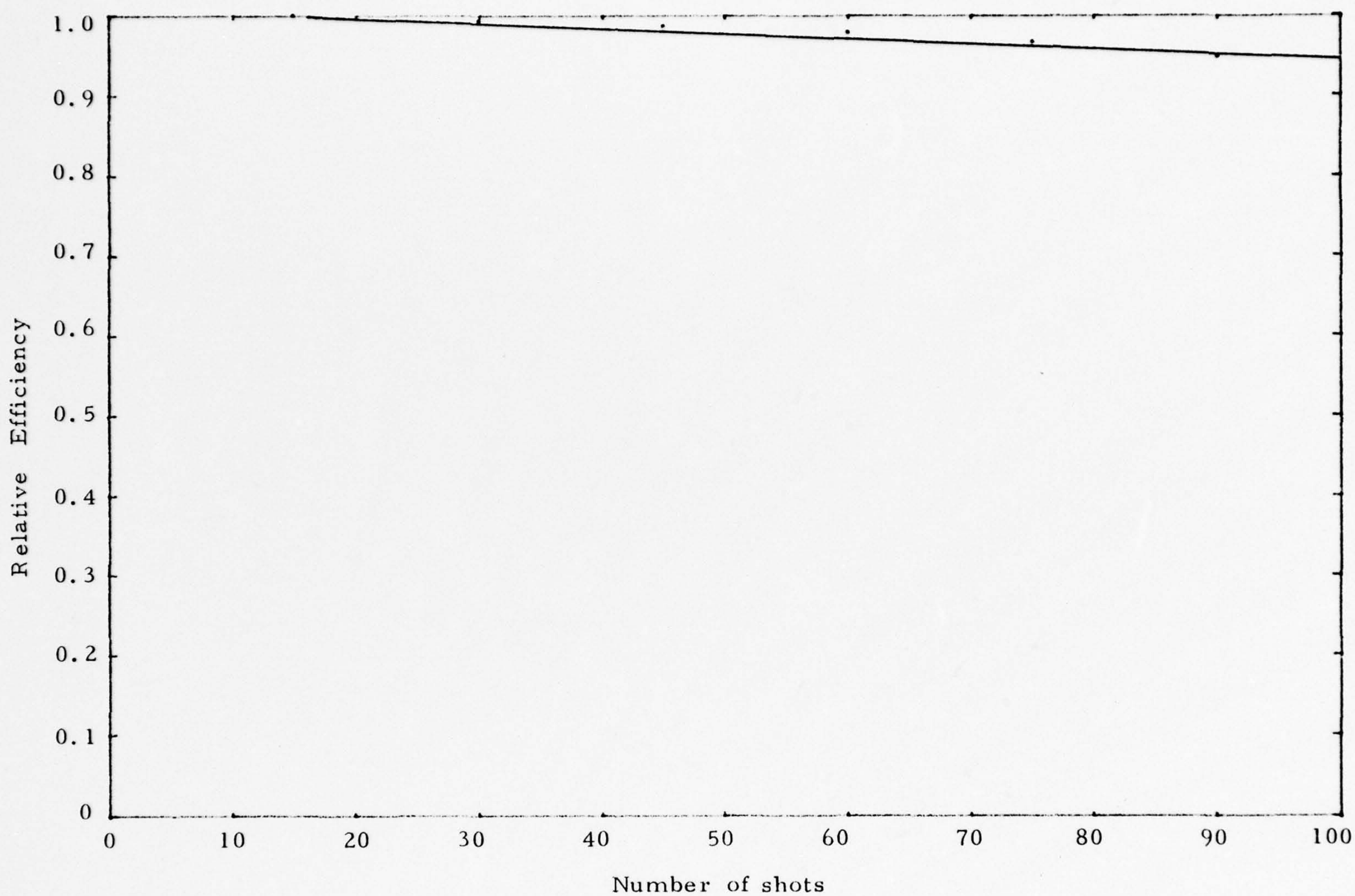

Figure 11. Stability of AC2F with KrF pumping. E_{in} = 90mJ/shot

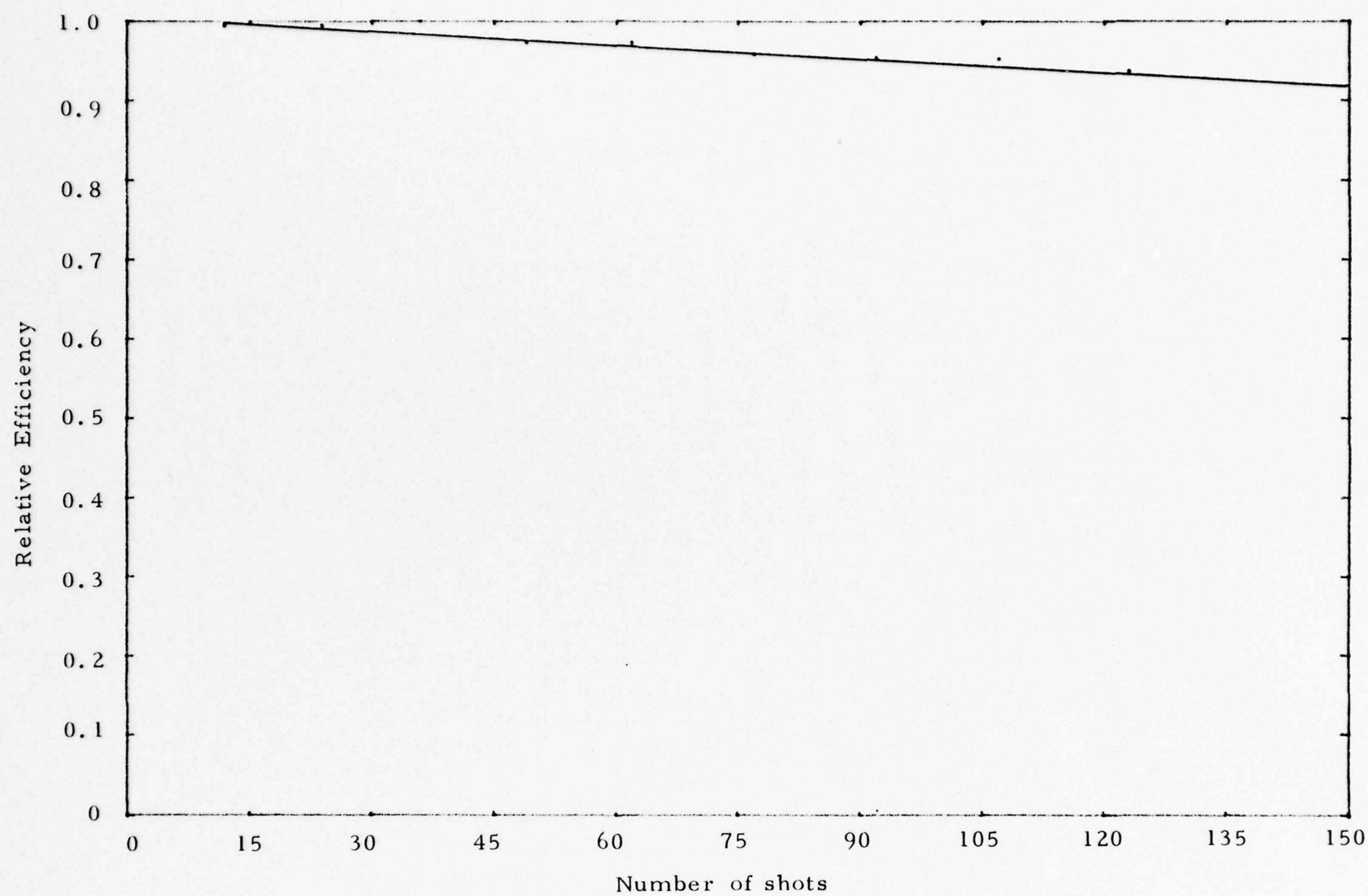

Figure 12. Stability of AC2F with KrF pumping. E_{in} = 90mJ/shot

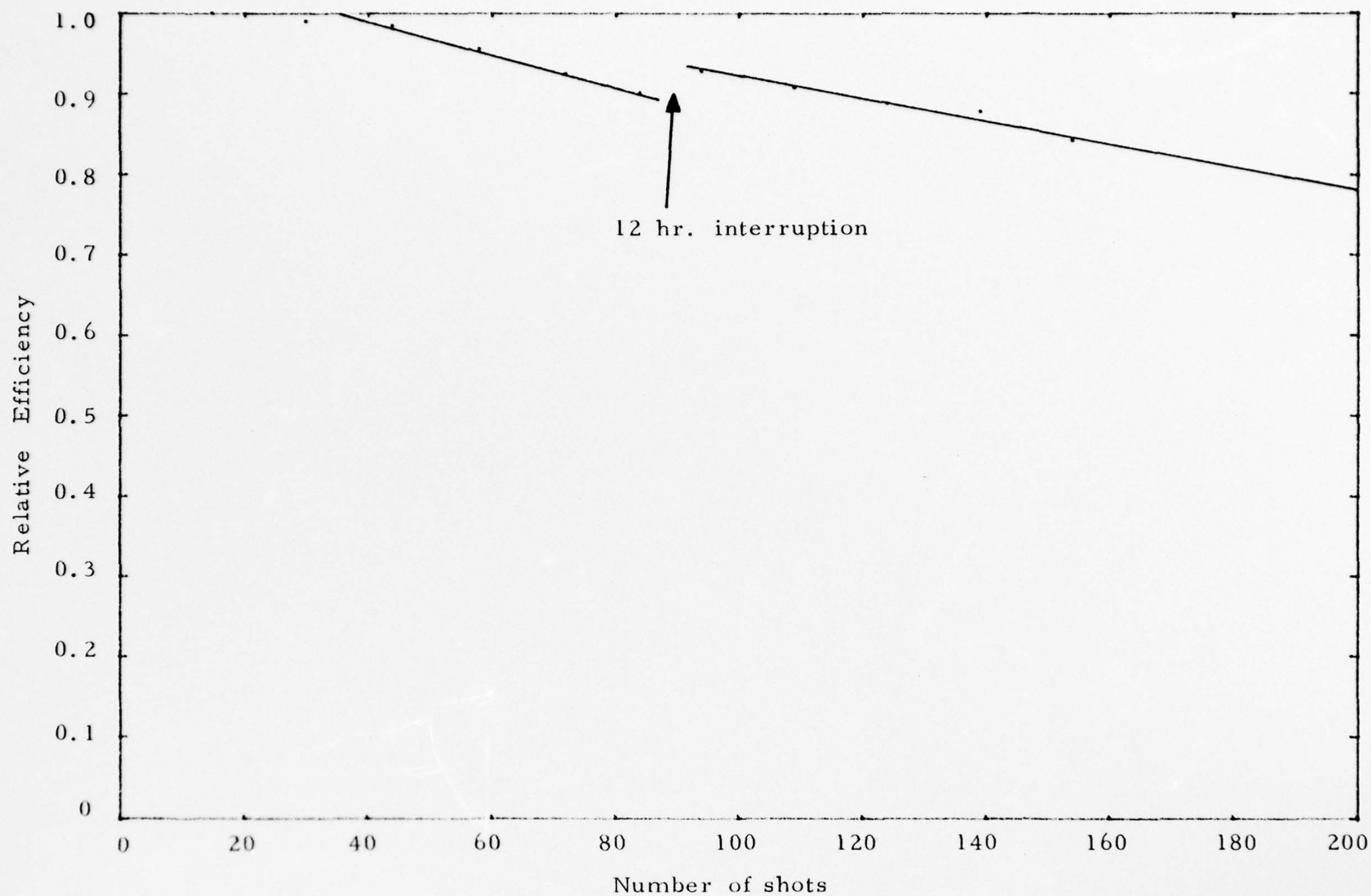

Figure 13. Stability of AC3F with KrF pumping. E_{in} = 90 mJ/shot

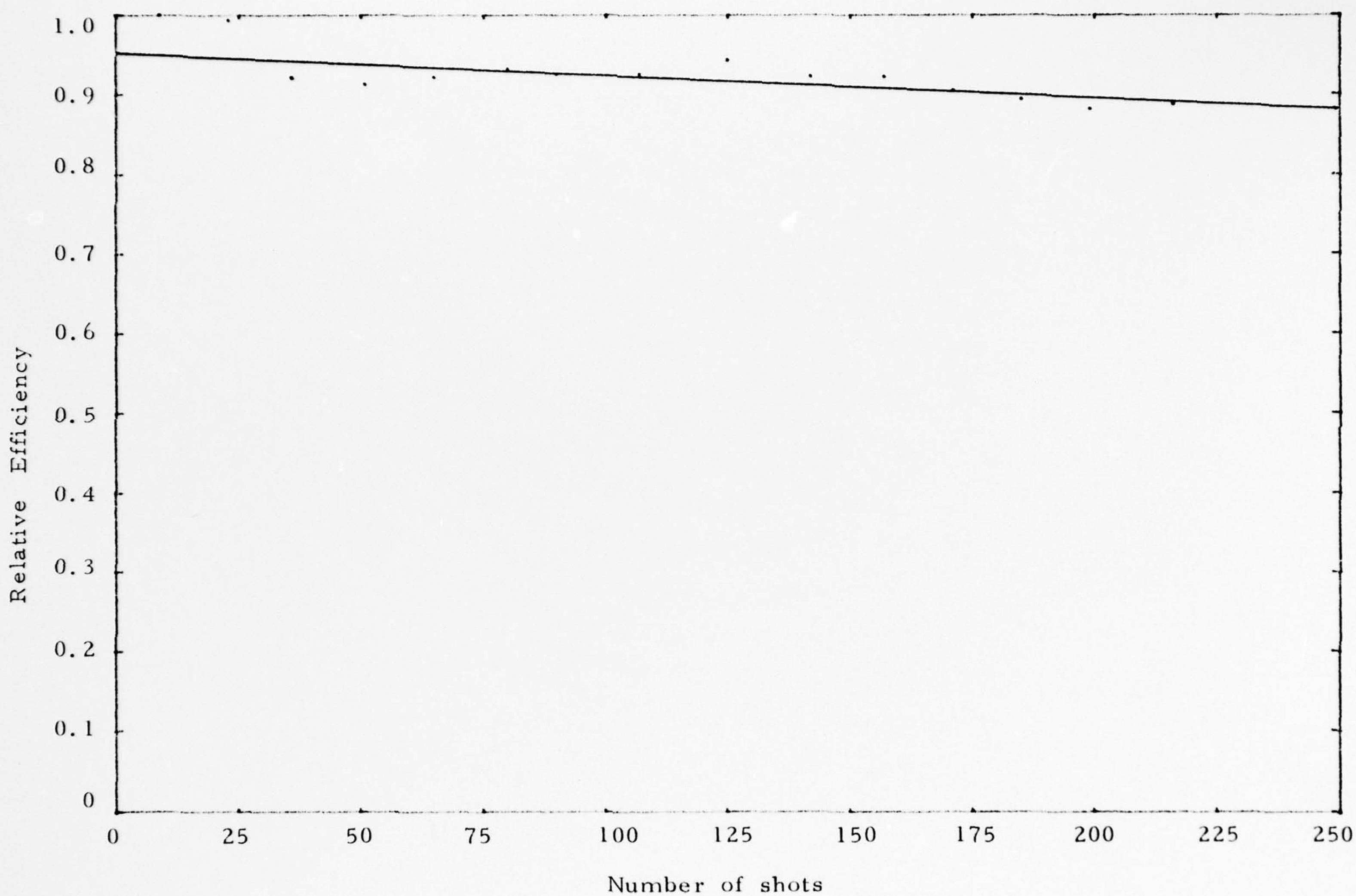

Figure 14. Stability of C102 with XeF pumping. $E_{in} = 50mJ/shot$

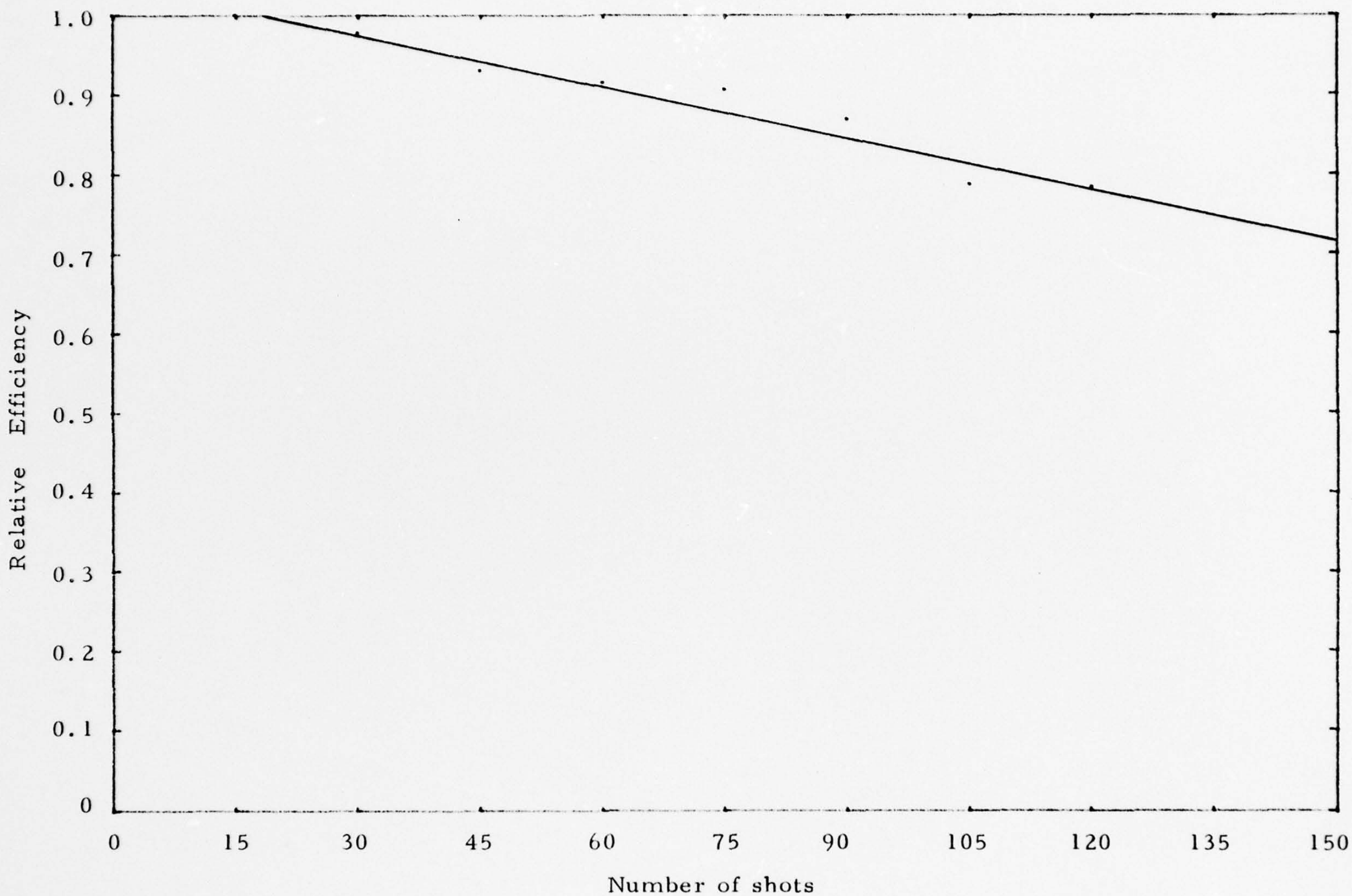

Figure 15. Stability of C2H with XeF pumping. E_{in} = 50mJ/shot

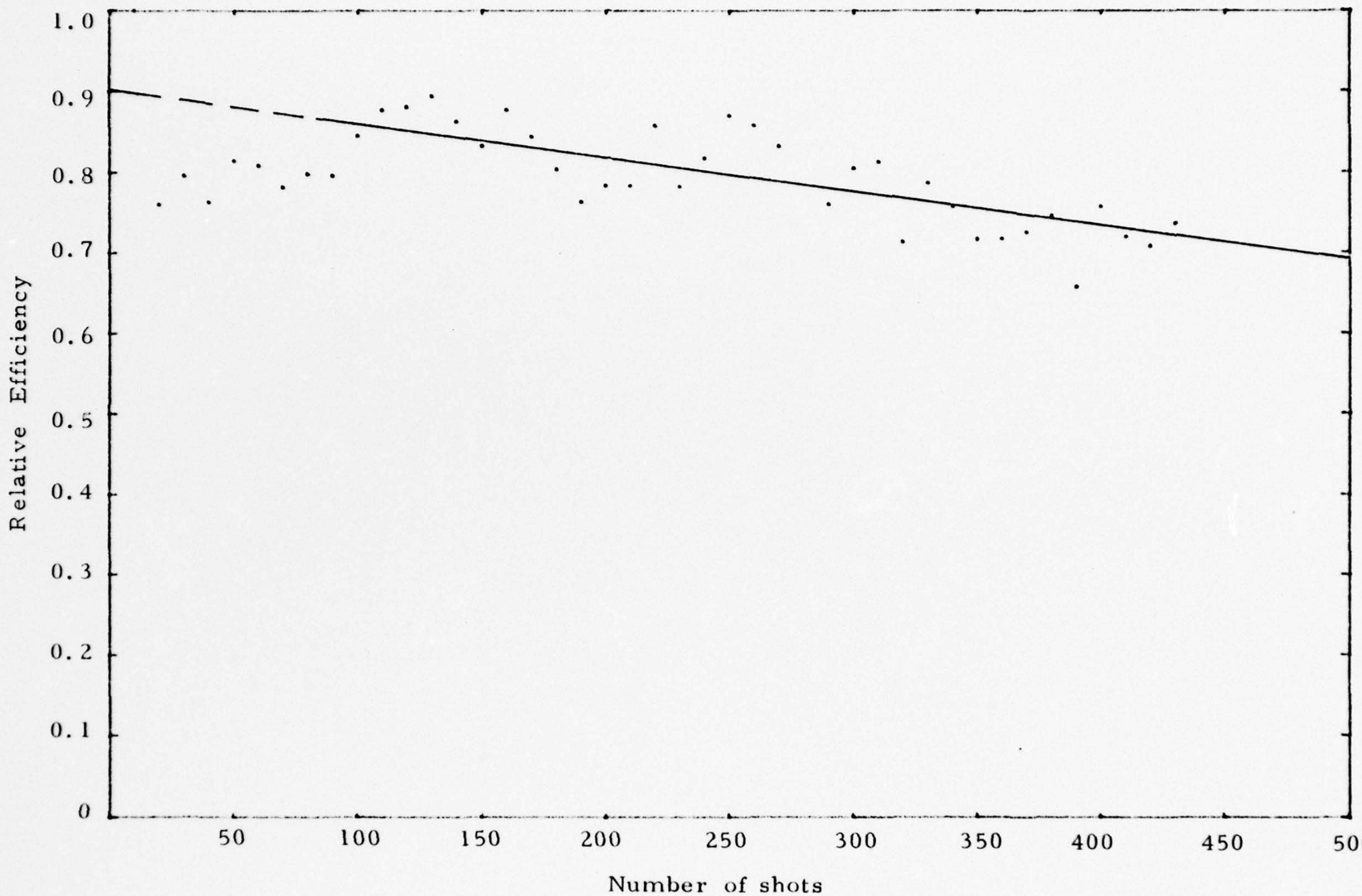

Figure 16. Stability of C6H with XeF pumping. $E_{in} = 50mJ/shot$

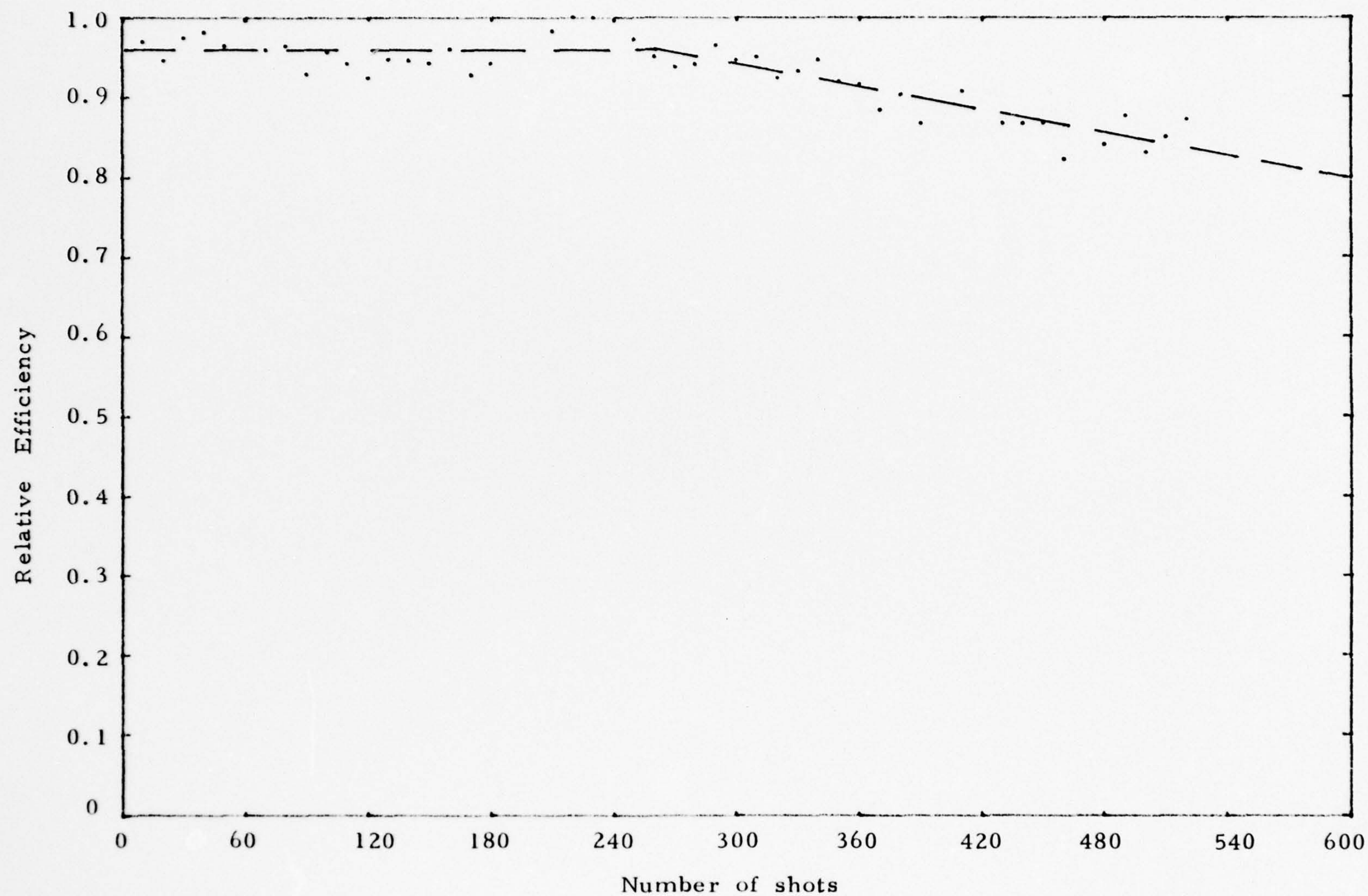

Figure 17. Stability of AC2F with XeF pumping. $E_{in} = 60mJ/shot$

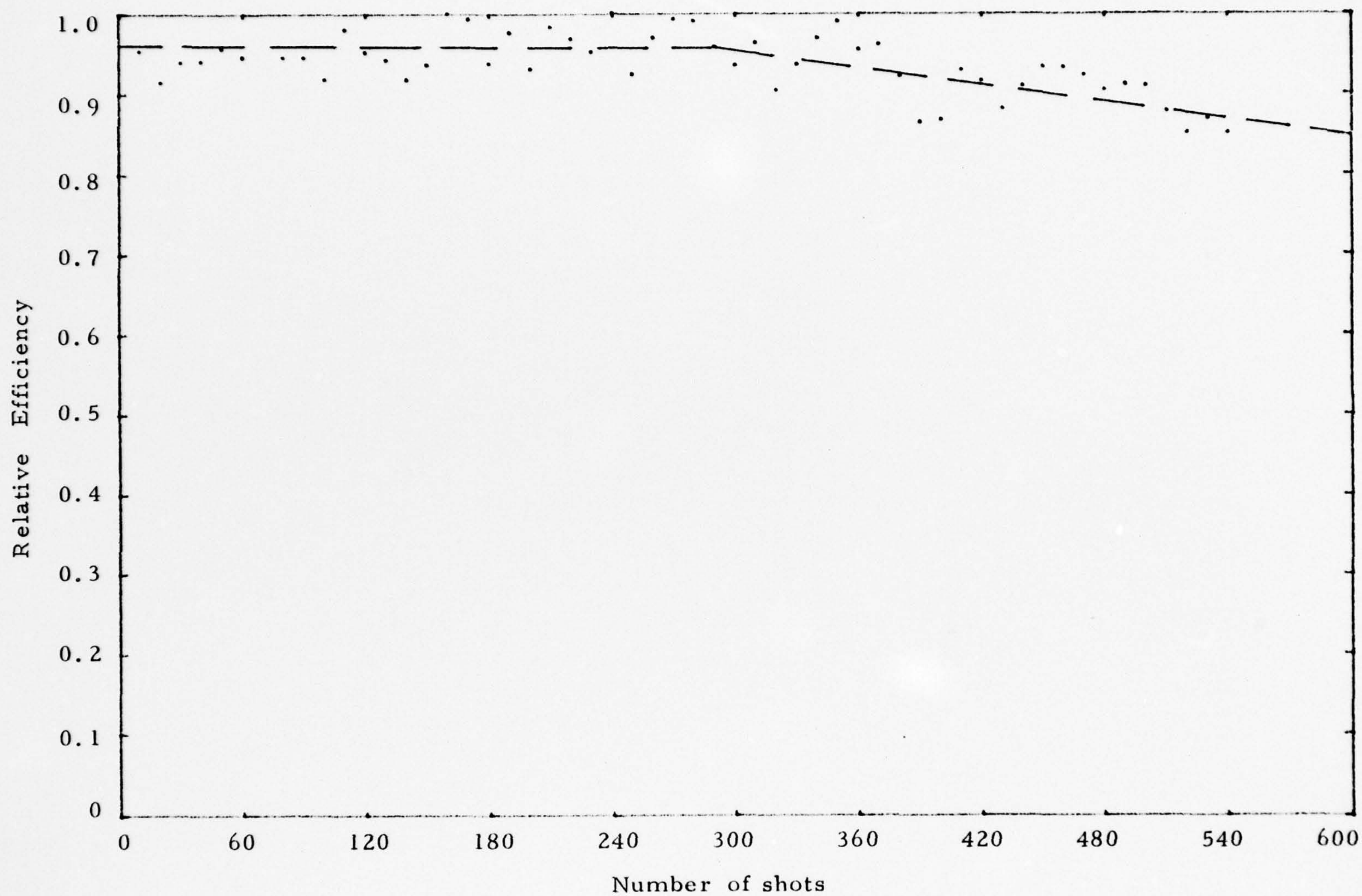

Figure 18. Stability of AC3F with XeF pumping. $E_{in} = 60 mJ/shot$

2.2 Stability and Efficiency at High Intensities

In addition to the stability measurements discussed in the previous section, the stability of the dye AC2F with KrF pumping was investigated at a pumping intensity about three times higher than the one used in the first series of tests, i.e., the uv energy was 270 mJ/shot instead of 90 mJ/shot. This particular dye was selected because of its high stability measured in the 90 mJ test. From Figure 19, $N_{1/2}$ is calculated to be 180, which has to be compared to 800 for the 90 mJ test. This decrease is somewhat faster than linear, i.e., a factor of four instead of three, but the difference is probably too small to be meaningful. Finally, Figure 20 shows the measured dependency of efficiency on pump intensity. At the highest intensity used in the test, the efficiency was still very close to its maximum value.

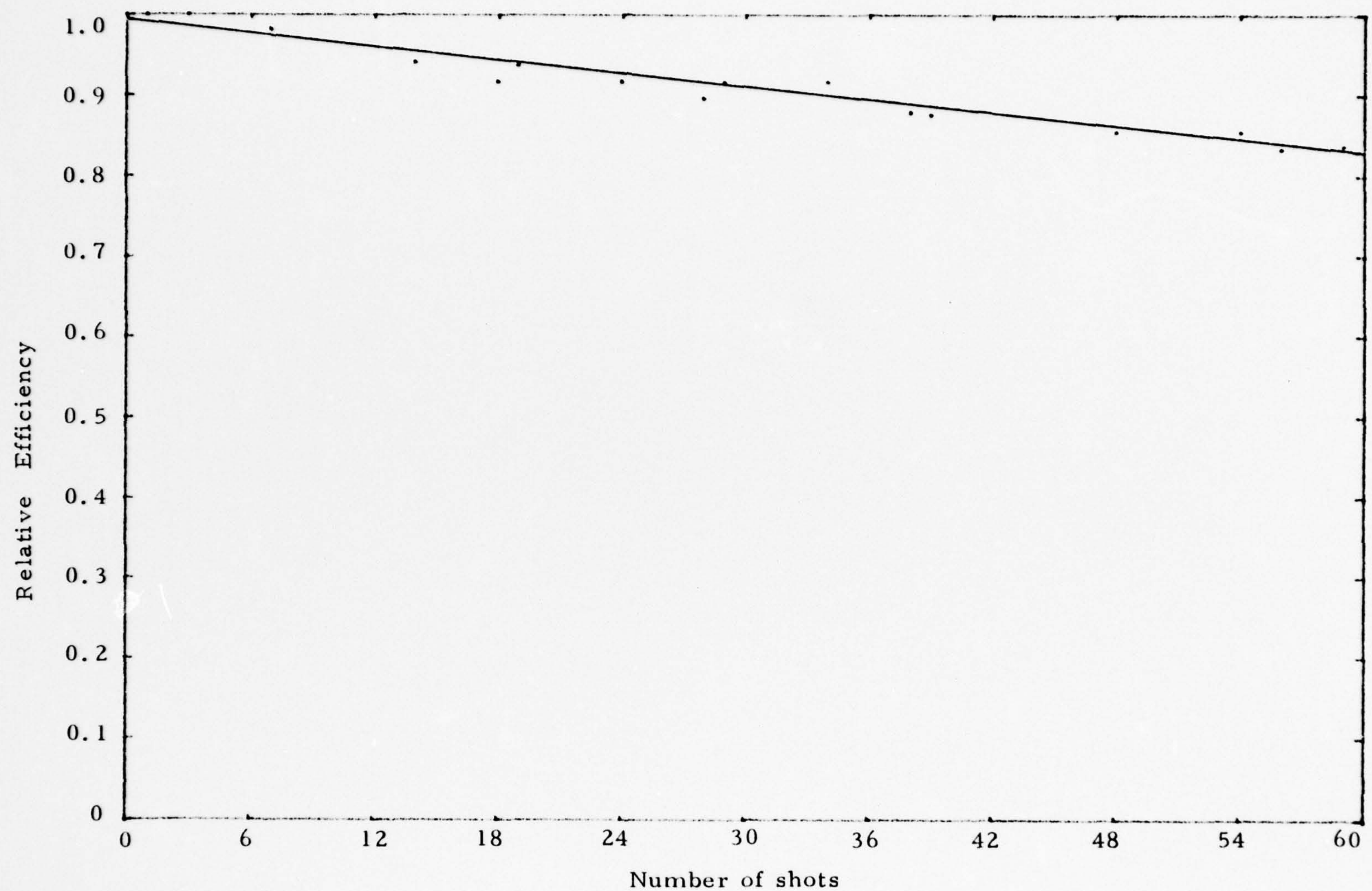

Figure 19. Stability of AC2F with KrF pumping. $E_{in} = 270mJ/shot$

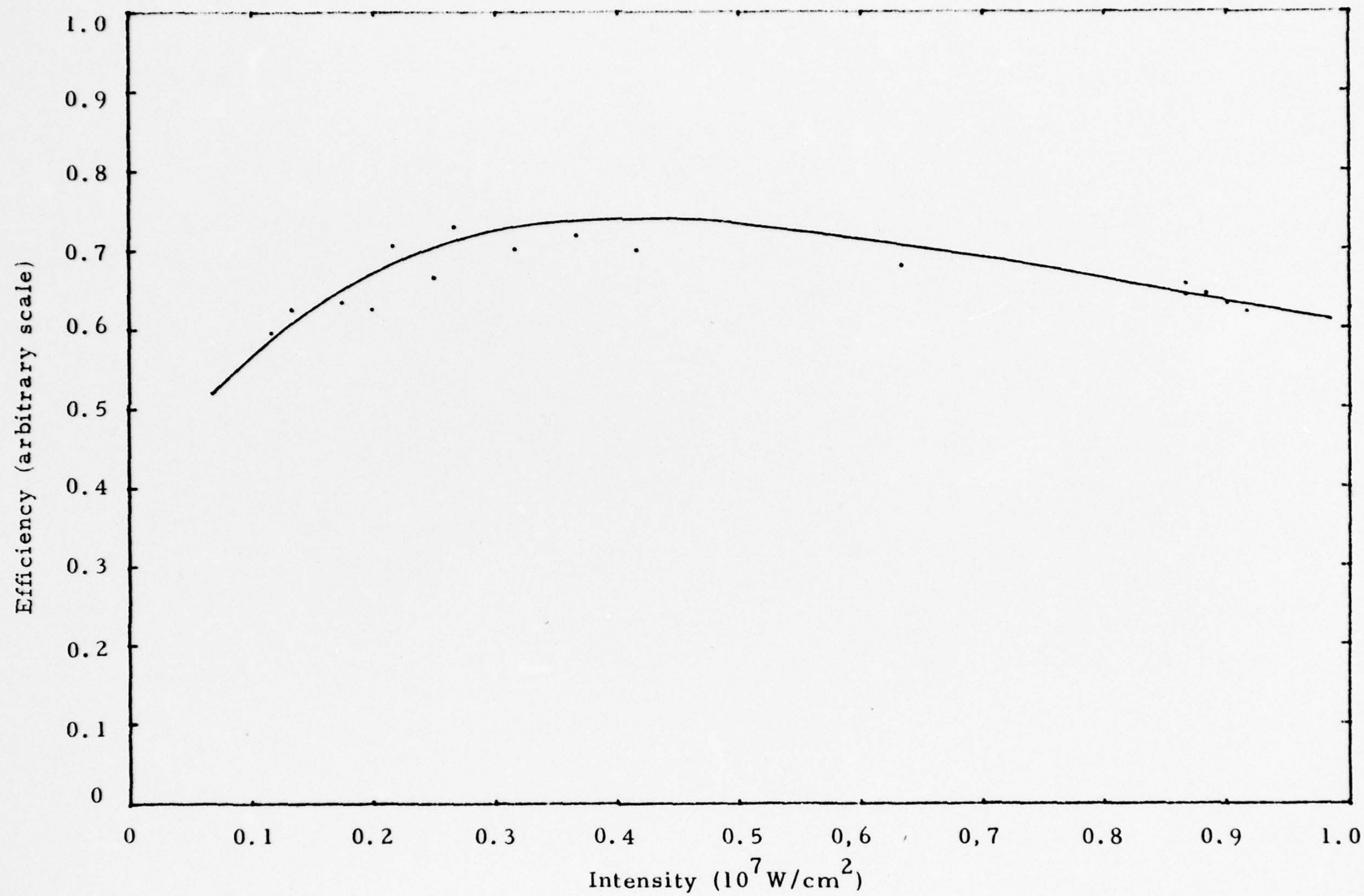

Figure 20. Efficiency versus pump intensity for AC2F with KrF pumping.

3.0 CONCLUSIONS

The photochemical stability of ten different dyes for uv pumping by KrF and XeF lasers has been investigated. Some of the dyes were found to be considerably more stable for laser pumping than for flash lamp pumping.[2] One reason for this increased stability is probably the shorter pulse length and faster rise time compared to flash lamp pumping, resulting in smaller populations in the reactive triplet states. Also, in the case of flash lamp pumping, part of the broadband flash lamp spectrum may photodecompose the dye molecules. The improved stability should make these dyes very attractive uv to blue-green frequency converters for underwater communication and for isotope separation.

ACKNOWLEDGEMENT

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2. E. J. Schimitschek, J. A. Trias, "New Laser Dyes with Blue-Green Emission," Optics Communications, Vol. 16, No. 3, March 1976.